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**PATENT APPLICATION**



**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of

Docket No: Q64848

Shinya WATANABE , et al.

Appln. No.: 09/875,158

Group Art Unit: 1616

Confirmation No.: 7273

Examiner: B. Badio

Filed: June 07, 2001

For: PRODUCTION PROCESS OF CYCLOHEXENYL METHYL KETONES

**DECLARATION UNDER 37 C.F.R. § 1.132**

Commissioner for Patents  
Washington, D.C. 20231

**RECEIVED**  
JUN 13 2002  
TECH CENTER 1600/2900

Sir:

I, Takeshi Yamamoto declare and state as follows:

I graduated from Shizuoka University, Faculty of Engineering, receiving a Master's Degree in March of 1969.

I have been employed by the Assignee of the present application, the Takasago International Corporation, since April of 1969.

From April of 1969 to the present, I have been engaged in the synthesis of conventional and novel perfume materials at the Central Research Laboratory of said company.

I am a co-inventor of the present application.

I reviewed the specification, the claims, all Official Actions and all prior art of record in the present application, and I'm well acquainted with all of that material, including U.S. Patent 4,334,098 Mookherjee et al (Mookherjee).

I have performed or have had performed under my direction and control certain experimentation regarding the present application and Mookherjee, which experimentation is set forth below.

#### EXPERIMENTATION

The process of the present invention and the process of Mookherjee are the same in that the  $\delta$ -trans isomer (1a') is obtained from  $\delta$ -cis isomer (1a). However, in Mookherjee the yield is 75% in Example II and 78% in Example X (1085 g to 847 g). Mookherjee does not describe the  $\alpha$ -isomer (1b) and  $\beta$ -isomer (1c) where the position of the double bond has been changed.

Mookherjee obtained  $\delta$ -trans isomer (1a') in Fractions 2-7 (b.p. 85 to 90°C, 9-11 mm pressure) in Example X. In addition, Fraction 1 (b.p. 38-81°C, 9 mm) pressure and Fraction 8 (b.p. 97°C, 11 mm pressure) were obtained. If the  $\alpha$ -isomer (1b) and  $\beta$ -isomer (1c) where the position of the double bond has been changed might be formed in the process of Mookherjee, the  $\alpha$ -isomer (1b) and  $\beta$ -isomer (1c) should exist in Fraction 1 or Fraction 8.

The relative volatilities of the  $\delta$ -cis isomer (1a), the  $\delta$ -trans isomer (1a'), the  $\alpha$ -isomer (1b) and the  $\beta$ -isomer (1c) were measured with the following results:

Average relative volatilities (according to Fenske's equation)

$\delta$ -trans isomer (1a')	$\alpha$ -isomer (1b)	$\delta$ -cis isomer (1a)	$\beta$ -isomer (1c)
1.1650	1.0000		
1.1765	1.0099	1.0000	
1.2956	1.1013	1.0905	1.0000

From the above table, it is clear that  $\delta$ -trans isomer (1a') has the highest volatility (i.e., the lowest boiling point). Thus, it was assumed there might be a possibility that  $\alpha$ -isomer (1b) and  $\beta$ -isomer (1c) exist in Fraction 8 of Mookherjee.

The following experimentation following the essential conditions of Mookherjee was then carried out. The results are also shown.

Catalyst	Temp./ Time	Solvent	1a	1a'	1b	1c
KOH (10 wt%)	135°C/4hr	Polythylene glycol dimethylether	6.7	93.3	0.0	0.0
28% NaOMe Methanol solution (10 wt%)	85°C/4 hr	Methanol	10.1	89.9	0.0	0.0

As is clear from the above table, it is not possible to obtain  $\alpha$ -isomer (1b) and  $\beta$ -isomer (1c) following the process of Mookherjee. Accordingly, Fraction 8 in Example X of Mookherjee contains the starting material,  $\delta$ -cis isomer (1a), not  $\alpha$ -isomer (1b) and  $\beta$ -isomer (1c).

The distinction, the object of the present invention is to provide the  $\alpha$ -isomer (1b) and  $\beta$ -isomer (1c) which cannot be obtained by the process of Mookherjee. Because the compounds obtained are different between the present invention and Mookherjee, the only logical conclusion is that the processes are different.

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The reaction in Example II of Mookherjee is in ethanol (b.p. 78°C) for 25 hours, and the reaction in Example X of Mookherjee is in methanol (b.p. 64°C) for 24 hours. Because these reactions were carried out under reflux, the boiling point of the solvent is almost the same as the reaction temperature.

In the present application, the reaction of Example 1 was at 90°C for 24 hours, the reaction of Example 2 was at 175 °C for 4 hours, the reaction of Example 3 was at 100°C for 4 hours, the reaction of Example 4 was at 175°C for 6 hours, the reaction of Example 5 was at 120°C for 2.5 hours, the reaction of Example 6 was at 140 to 145°C for 5 hours, the reaction of Example 7 was at 120°C for 4 hours, the reaction of Example 8 was at 130 to 140°C for 6 hours, the reaction of Example 9 was at 140 to 145°C for 6 hours, the reaction of Example 10 was at 170 to 190°C for 6 hours, and the reaction of Example 11 was at 135°C for 3 hours.

In Mookherjee, the reaction is carried out under conventional basic conditions in an alcoholic solvent, i.e., Example II uses potassium hydroxide as a base in methanol and Example X uses sodium methoxide as a base in methanol.

In distinction, when a base is used in the present invention, use of a solvent having a high boiling point and high polarity such as tetraethyleneglycol monomethyl ether (Example 2), dimethyl sulfoxide (Examples 3, 5, 6, 7, 8, 10), dimethylacetamide (Example 9), or cyclohexylamine (Example 11) enhances the intensity of the function of the base used in the reaction system so that the reaction can proceed under strong basic conditions. Accordingly, the

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reaction of the present invention proceeds under much stronger basic conditions than Mookherjee.

Further, according to the present invention, an acid may be used in the reaction (Examples 1 and 4). Mookherjee does not teach or suggest reaction under acidic conditions.

As a result, the process of the present invention can provide 32.5% or more of double-bond isomers ( $\alpha$ -isomer (1b) and  $\beta$ -isomer (1c)) (cf. Example 10).

In particular, the present invention permits obtaining 47.7% or more of double-bond isomers ( $\alpha$ -isomer (1b) and  $\beta$ -isomer (1c)) (cf. Example 7) by using potassium t-butoxide (Examples 2, 3, 6, 7, 9) having higher base city than potassium hydroxide and sodium methoxide, as used in Mookherjee.

As it is clear from the above explanation, the conditions for the process of the present invention are quite different from those of Mookherjee. The process of the present invention enables one to obtain  $\alpha$ -isomer (1b) and  $\beta$ -isomer (1c) which cannot be obtained by the process of Mookherjee.

DECLARATION UNDER 37 C.F.R. § 1.132  
U.S. Application No.: 09/875,158

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: \_\_\_\_\_

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Takeshi Yamamoto

02- 6-12:12:18 : 高橋和生 (長) 研究企画部

法務特許部

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FROM SUGHRUE MION WASHINGTON DC

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PATENT APPLICATION

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

Docket No: Q64348

Shinya WATANABE, et al

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Filed: June 07, 2001

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## DECLARATION UNDER 37 C.F.R. § 1.132

Commissioner for Patents  
Washington, D.C. 20231

Sir:

I, Takeshi Yamamoto declare and state as follows:

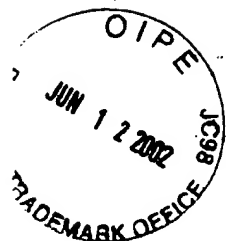
I graduated from Shizuoka University, Faculty of Engineering, receiving a Master's Degree in March of 1969.

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I am a co-inventor of the present application.

I reviewed the specification, the claims, all Official Actions and all prior art of record in the present application, and I'm well acquainted with all of that material, including U.S. Patent 4,334,098 Mookherjee et al (Mookherjee).



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02- 6-12:12:18 :商務部工業(株)研究本部

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DECLARATION UNDER 37 C.F.R. § 1.132  
U.S. Application No.: 09/875,158

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02- 6-12:12:18 :高砂香料工業(株) 研究企画部

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DECLARATION UNDER 37 C.F.R. § 1.132  
U.S. Application No.: 09/875,158

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02- 6-12:12:18 : 富時信託工業 (株) 研究企画部

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DECLARATION UNDER 37 C.F.R. § 1.132  
U.S. Application No.: 09/875,158

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DECLARATION UNDER 37 C.F.R. § 1.132  
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FROM SUGHRUE MION WASHINGTON DC

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DECLARATION UNDER 37 C.F.R. § 1.132  
U.S. Application No.: 09/875,158

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: June 12, 2002

Takeshi Yamamoto  
Takeshi Yamamoto